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A fractal model for predicting the effective thermal conductivity of liquid with suspension of nanoparticles

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Abstract

Based on the effective medium approximation and the fractal theory for the description of nanoparticle cluster and its radial distribution, a method for modeling the effective thermal conductivity of "nanofluid" is established. The size effect and the surface adsorption of nanoparticles are taken into considerations. The proposed fractal model is discussed in detail for its application, and it predicts quite well with our recent measuring data for dilute suspensions of metallic oxide nanoparticles.

Keywords: Effective thermal conductivity; Nanoparticles; Clustering; Fractal model; Surface adsorption; Size effect

1. Introduction

The researches on the effective thermal conductivity of liquid with nanoparticle inclusions attract more and more interests experimentally and theoretically. The effective thermal conductivity of nanoparticle suspension can be much higher than the normally used industrial heat transfer fluid, such a fluid has terminologized as "nanofluid" by S.U.-S. Choi of Argonne National Laboratory of USA in 1995, and considered to be a novel enhanced heat transfer fluid. Very recently, Keblinski et al. [1] reported their idea on the possible mechanisms of enhancing thermal conductivity, and suggested that the size effect, the clustering of nanoparticles and the surface adsorption could be the major reason of enhancement, while the Brownian motion of nanoparticles contributes much less than other factors. Wang and Peng [2] have studied experimentally the effective thermal conductivity of liquids with 25 nm SiO_2 particle inclusions, and observed the percolation pattern of particle clustering by scanning tunnel microscopic (STM) photos. It was believed that clustering could affect the enhancement prominently. As the measurement is made by an unsteady thermal-probe method, the effect of liquid convection cannot be avoided. Thus, a novel measurement method, named as quasi-steady-state method and is usually used in the measurements of thermophysical properties of solids, was adapted for new measurements, to exclude the effect of local convection [3]. The same as we reported previously, the modeling of effective thermal conductivity of nanoparticle suspension including the effect of clustering would be necessary.

The fractal theory was proposed firstly by Mandelbrot [4], a French mathematician. It can well describe the disorder and stochastic process of clustering and polarization of nanoparticles within the mesoscale limit. Pitchumani and Yao [5] have firstly used fractal theory in the research of effective thermal conductivity for unidirectional fibrous composites, and obtained their fractal characters. Yu et al. [6–8] obtained a fractal description of effective dielectric coefficient of composite material using the traditional effective medium theory and the widely used fractal theory. But, few reports to use the fractal theory in descripting the cluster of nanoparticle suspensions to predict the effective thermal conductivity.

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Nomenclature

a	radius of nanoparticles
D_f	fractal dimension
D_{f1}	fractal dimension of clusters
D_{f2}	fractal dimension of cluster distribution
D_w	anomalous diffusive index
D_s	fractal sub-dimension
k	effective thermal conductivity
l	mean free path of phonons
M	molecular weight of liquid
$n(r)$	radius distribution function
N_A	Avogadro constant
q	heat flux
r	equivalent radius of cluster
t	thickness of adsorption monolayer
T	temperature
ΔT	temperature difference

Greek symbols

ϕ	volume fraction
δ	thickness of sample

Subscripts

ad	adsorption
b	bulk
cl	cluster
eff	effective
f	liquid
p	(nano)particle

In this paper, we will introduce briefly the effective medium theory and the concept of fractal dimension for nanoparticle clusters, together with the space description of cluster radius, considering the effect of particle size and surface adsorption, and attempt to establish a fractal model for predicting the effective thermal conductivity of liquid with nanoparticle inclusion.

2. Effective medium theory

There are two methods commonly used in effective medium theory to treat the effective transport coefficient of mixture and composites: the Maxwell–Garnett's self-consistent approximation (MG model) [9] and the Bruggeman approach [10]. The former one fits well with experimental data for dilute and randomly distributed components included in a homogeneous host medium, the particles are considered as to be isolated in the host medium, no interactions existing among them. For the two-component entity of spherical-particle suspensions, the MG model [9] gives

$$\frac{k_{\text{eff}}}{k_f} = \frac{(1-\phi)(k_p + 2k_f) + 3\phi k_p}{(1-\phi)(k_p + 2k_f) + 3\phi k_f}, \tag{1}$$

where k_{eff} is the effective thermal conductivity of liquid with particle suspension, k_f the thermal conductivity of host medium, k_p the thermal conductivity of particle, and ϕ the volume fraction of particles. The MG model is applicable to suspension with low-concentration particle inclusions.

The Bruggeman model with mean field approach is used to analysis the interactions among the randomly distributed. For a binary mixture of homogeneous spherical inclusions, the Bruggeman model [10] gives

$$\phi\left(\frac{k_p - k_{\text{eff}}}{k_p + 2k_{\text{eff}}}\right) + (1-\phi)\left(\frac{k_f - k_{\text{eff}}}{k_f + 2k_{\text{eff}}}\right) = 0 \tag{2}$$

and the solution of above quadratic equation is given as:

$$k_{\text{eff}} = (3\phi - 1)k_p + [3(1-\phi) - 1)]k_f + \sqrt{\Delta}, \tag{3}$$

$$\Delta = (3\phi - 1)^2 k_p^2 + [3(1-\phi) - 1)]^2 k_f^2 + 2[2 + 9\phi(1-\phi)]k_p k_f. \tag{4}$$

The Bruggeman model has no limitation on the concentration of inclusions, and can be used for particle percolation in suspensions.

For low particle-concentration suspension, the Bruggeman model shows almost the same result as the MG model will give. For a particle percolation situation or when the particle concentration is sufficiently high, the MG model fails to predict precisely the experimental results, while the Bruggeman model can still fit well with experimental data. Hence, we will use Bruggeman model to predict the effective thermal conductivity of nanoparticle clustering, and use otherwise the MG model to approximate the effective thermal conductivity of nanoparticle suspensions.

3. Fractal indexes

As Yu et al. [6–8] have proposed, we will use Bruggeman model and fractal theory to predict the effective thermal conductivity of nanoparticle clusters. In the aspect of fractals, Havlin and Ben-Avraham [11] figured out that, the radius distribution of nanoparticles and the distribution of nanoparticles in suspension have both shown some kind of self-comparability. The scaling theory is commonly used for the quantitative description

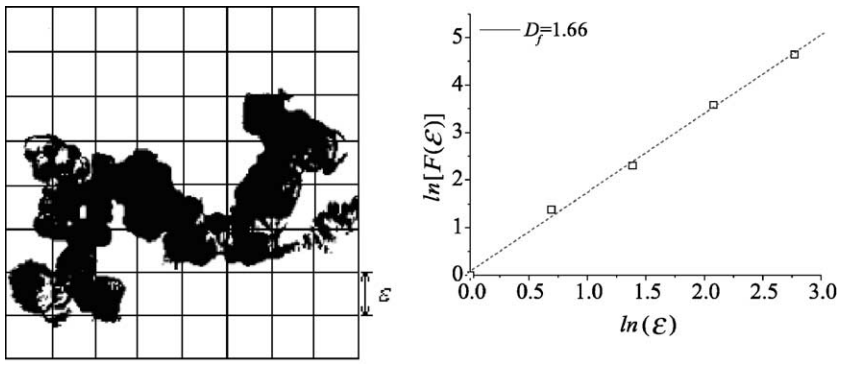

Fig. 1. The fractal dimension of section area of SiO_2/ethanol cluster.

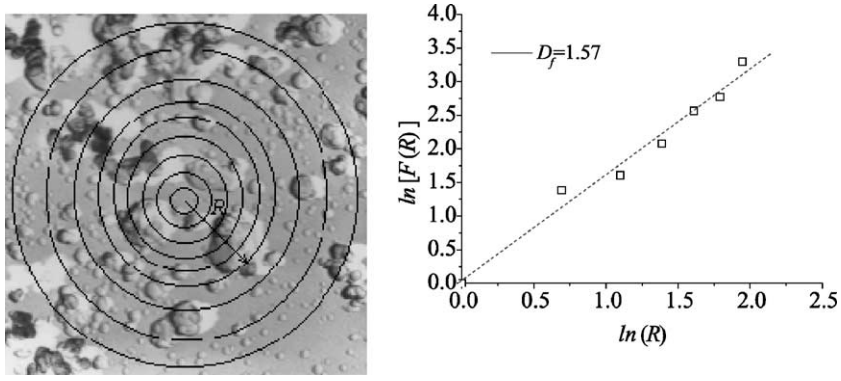

Fig. 2. The fractal dimension of radius distribution of clusters.

of fractal system. But, before going forward for its actual use, it is needed to introduce some definitions of fractal indexes.

The fractal dimension, D_f, is one of the basic variables for the description of fractals. It is established through a scalar with unit ε. If the volume (area, particle numbers, etc.) of the fractal is $F(\varepsilon)$, then, the fractal dimension D_f can be decided through the following expression:

$$F(\varepsilon) = C\varepsilon^{D_f}, \tag{5}$$

where C is a shape factor that is independent of ε. Different fractal indexes are needed when describing the complex fractals. The "anomalous diffusive index", D_w, reflects the self-comparability of particle diffusion in the host system, and can be approximated by the following relation between D_w and D_f:

$$2D_f/D_w = D_s \doteq 1.33, \tag{6}$$

where D_s is the fractal sub-dimension that stands for the self-comparability of space density fractals. The anomalous diffusive index, D_w, can thus be determined with calculated fractal dimension, D_f.

It is necessary to note that the fractal dimension of clusters, D_{f1}, is different from that for space distribution of clusters, D_{f2}. Both of them can be determined through experiments. For particles of 25 nm SiO_2 (mass concentration, 6.5%) suspended in ethanol (purity, 99.7%), the electron microscopic photos of clusters in suspension, Fig. 1, and the radius distribution of them, Fig. 2, can be taken by corresponding technology, with thin film prepared by the quick freezing method of liquid helium [2]. In Fig. 1, the fractal dimension of clusters, D_{f1}, is calculated to be 1.66; while the fractal dimension of space distribution of clusters, D_{f2}, is calculated to be 1.57.

4. Fractal model proposed

The enhancement in effective thermal conductivity for liquid with nano-sized particles relates directly with the particle interaction and clustering process. The nanoparticle suspension should be considered to be composed by host liquid and percolation patterned cluster inclusions. Thus, when using Eq. (1), k_p will be replaced by the effective thermal conductivity of nanoparticle clusters, $k_{cl}(r)$, predicted by Bruggeman model.

Provided that different sizes of clusters, r, have formed in suspension due to the interaction of nanoparticles

of equal radius, a. The following equation can be obtained from the fractal theory [12,13]:

$$f(r) = (r/a)^{D_{\mathrm{fl}}-3}, \tag{7}$$

where r is the radius of nanoparticle clusters, $f(r)$ the volume fraction, and D_{fl} the fractal dimension. By Bruggeman approach [10], substitute $f(r)$ for ϕ into Eqs. (3) and (4), the effective thermal conductivity of cluster can be expressed as $k_{\mathrm{cl}} = k_{\mathrm{cl}}(r)$.

The suspension of particles with same radii, a, can now be treated as a suspension of clusters with different radius, r. Then, using a group of established fractal indexes, e.g. D_{f}, D_{w}, etc., the fractal characteristic of the space distribution of clusters can be described as [14]:

$$n(x) = Bx^g \exp(-bx^j), \tag{8}$$

where $x = r/n_\tau^{1/D_{\mathrm{w}}}$, B and b being, respectively, functions of randomly walk step number (time), n_τ, and g and j corresponding to different fractal indexes. So, the cluster size distribution varies with different n_τ. However, many experimental data have shown that different n_τ lead the function $n(r)$ to a consistent curve. To the classical Rosin–Rammlar fractal distribution function, the indexes are settled as: $j = D_{\mathrm{w}}/(D_{\mathrm{w}}-1)$, $g = (j-1)$. An alternative method, which describes the completely stochastic walk of a large amount of particles to form the disordered clusters through the short distance adherence forces, is the log normal distribution function. When the volume of particles can be expressed as $V = Hr^m$, in which H and m are constants about the shape factors of particles, the following log normal distribution function can approximately be used to describe $n(r)$ [15]:

$$n(r) = \frac{1}{r\sqrt{2\pi}\ln\sigma}\exp\left\{-\left[\frac{\ln(r/\bar{r})}{\sqrt{2\pi}\ln\sigma}\right]^2\right\}, \tag{9}$$

where $\bar{r}$ is the geometric mean radius, σ is the standard deviation. The value of $\bar{r}$ can be substituted approximately with the average radii, a, and σ can take the classic value of 1.5.

Using the multi-component MG model proposed by Wood and Ashcroft [16], we can obtain the effective thermal conductivity of suspension with nanoparticle inclusions, considering the effect of particle clustering and cluster distribution. Substitute the effective thermal conductivity of clusters, $k_{\mathrm{cl}}(r)$, and the radius distribution function, $n(r)$, into the modified MG equation, the effective thermal conductivity of nanoparticle suspension can be expressed as:

$$\frac{k_{\mathrm{eff}}}{k_{\mathrm{f}}} = \frac{(1-\phi)+3\phi\int_0^\infty \frac{k_{\mathrm{cl}}(r)n(r)}{k_{\mathrm{cl}}(r)+2k_{\mathrm{f}}}\,\mathrm{d}r}{(1-\phi)+3\phi\int_0^\infty \frac{k_{\mathrm{f}}n(r)}{k_{\mathrm{cl}}(r)+2k_{\mathrm{f}}}\,\mathrm{d}r}. \tag{10}$$

This equation is the proposed fractal model deduced for predicting of effective thermal conductivity of liquid with nanoparticles inclusion.

5. Consideration of size effect and surface adsorption of nanoparticles

Without consideration of radiation, the heat carriers in nanoparticles include only phonons and electrons. Chen [17] established the transport regimes for these heat carriers, according to the relation between the mean free path of carriers and the length scale of nanostructures. When the mean free path of heat carriers is comparable with the size of nanoparticles, i.e., 10–100 nm, the Boltzmann equation could be applicable for describing the heat transfer process. Hence, using the relaxation time approximation method [18], the effective thermal conductivity of nonmetallic nanoparticles can be approximated as

$$k_{\mathrm{p}} = \frac{3a^*/4}{3a^*/4+1}k_{\mathrm{b}}, \tag{11}$$

where k_{b} is the (bulk) thermal conductivity of particle, $a^* = a/l$ is the nondimensional radius, and l is the mean free path of phonons. For the metallic nanoparticles, the effective thermal conductivity can be achieved, provided that the Wiedemann–Franz Law still holds when the temperature is much higher than the Debye temperature. The size effect on the phonon–electron coupling factor is also negligible within the above-mentioned regime [19]. A cubic decreasing law was found in the effective electric conductivity for particles smaller than 500 nm [20]. Thus, when the relaxation times of electron and phonon are comparable, the following equation can be used for effective thermal conductivity of metallic nanoparticles:

$$k_{\mathrm{p}} = \left(\frac{2a}{5\times10^{-6}}\right)^3 k_{\mathrm{b}}. \tag{12}$$

Now, we take insights into surface adsorption. The adsorption of liquid molecules on the particle surface is thought to be a monolayer one. The way of molecule allocation on the surface is commonly considered to be a hexagonal closed-packed style. From the Langmuir formula of monolayer adsorption of molecules, the thickness of the adsorption layer can be expressed as [21].

$$t = \frac{1}{\sqrt{3}}\left(\frac{4M}{\rho_{\mathrm{f}}N_{\mathrm{A}}}\right)^{1/3}, \tag{13}$$

where M is the molecular weight of liquid, ρ_{f} is the density of liquid, and N_{A} is Avogadro constant (6.023×10^{23}/mol). Since the monolayer always occurs in conjunction with the particle sphere, they are completely correlated [22], and hence, the effective thermal con-

ductivity of the nanoparticle can be considered to be the total thermal conductivity of these two substances [16]:

$$k_{cp} = k_{ad}\frac{(k_p + 2k_{ad}) + 2A^3(k_p - k_{ad})}{(k_p + 2k_{ad}) - A^3(k_p - k_{ad})}, \quad (14)$$

where $A = 1 - t/(t + a)$, k_{ad} is the effective thermal conductivity of the adsorption layer. With the consideration of surface adsorption, we should substitute $(a + t)$, $[(a + t)/a]^3\phi$ and k_{cp} for a, ϕ and k_p, respectively, in Eqs. (2)–(10). The value of k_{ad} is hard to be predicted, but from Eq. (14), we can take $k_{ad} \to k_{cp}$ as first approximation, and thus the calculated results will stand for the upper bound of enhancement for effective thermal conductivity of liquid with nanoparticles inclusion.

6. Analysis and discussion

The three-component core-shell-medium (CSM) model [23] deduced from the MG approximation has considered the adsorption process on the particle surface. The Rayleigh model [24] concern the effect of

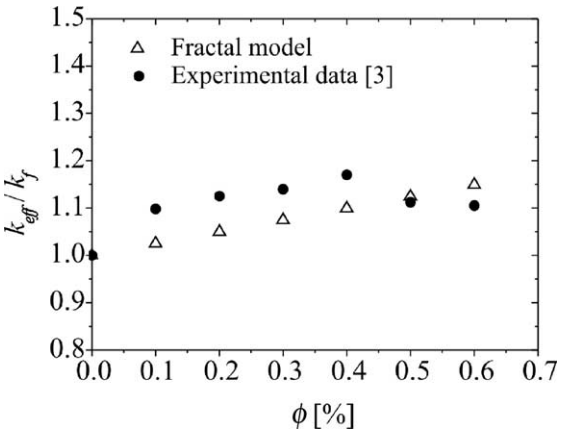

Fig. 4. Comparison of proposed fractal model with experimental data for CuO/deionized water.

Table 1
Comparison of the calculated value of k_{eff}/k_f using various models

Particle volume fraction (%)	Bruggeman model [10]	CSM model [23]	Rayleigh model [24]	CF model [25]	Monecke model [26]	Experimental results [3]
0.1	1.00262	1.00192	1.00262	1.00266	1.00262	1.0982
0.2	1.00526	1.00386	1.00524	1.00539	1.00525	1.1252
0.3	1.00791	1.00582	1.00787	1.0082	1.00788	1.13984
0.4	1.01057	1.00781	1.01051	1.01108	1.01051	1.16996
0.5	1.01324	1.00982	1.01314	1.01404	1.01316	1.11238
0.6	1.01593	1.01185	1.01579	1.01708	1.01581	1.10531

Table 2
Data for calculation

Silicon dioxide		Ethanol	
Average radius	$a = 25$ nm		
Mean free path of phonons	$l = 14$ nm	Thickness of adsorption monolayer	$t = 2.8$ nm
Density	$\rho_p = 6310$ kg/m^3	Density	$\rho_f = 996$ kg/m^3
Thermal conductivity	$k_p = 32.9$ W/m/K	Thermal conductivity	$k_f = 0.613$ W/m/K

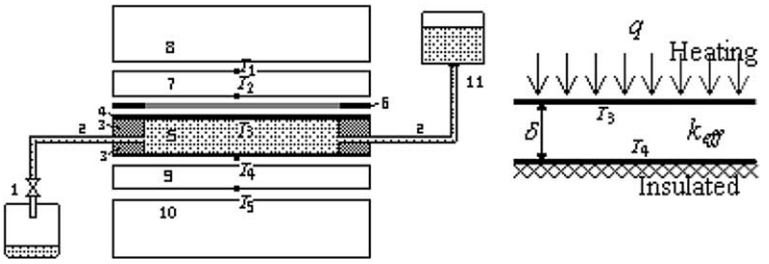

Fig. 3. Measuring apparatus. (1) Valve; (2) ducting tube; (3) supporter (insulator); (4) Al sheet; (5) sample liquid; (6) plane heater; (7,9) heat-loss measuring layer; (8,10) insulator; (11) reservoir.

particle interaction, but for particle of small radius, its accuracy is relatively higher than the MG model. The Cichocki–Felderhof (CF) model [25] came from statistical method and considered the interaction between particles of same radius. The Monecke model [26] discarded the physical topology technique of effective medium theory, deduced on the assumption that the effective thermal conductivity equals to an interpolation between the extreme limits of its components. We compare these models with experimental results for suspension of CuO nanoparticles (50 nm) in deionized water [3] in Table 1. Data used for calculation are listed in Table 2. All these models function as the same in dilute limit, yet none of them explains well with our experimental data [3].

Our experiments were conducted on an apparatus shown in Fig. 3, which was specially designed to suit the condition, for which the testing medium is kept in its original uniform temperature, T_0 before being heated, and the analytical solution is given by Carslaw and Jaeger [27] as

$$k_{\text{eff}} = q\delta/(2\Delta T), \tag{15}$$

where q is the constant heat flux from the heating surface, δ is the thickness of sample, $\Delta T = (T_3 - T_4)$ is the

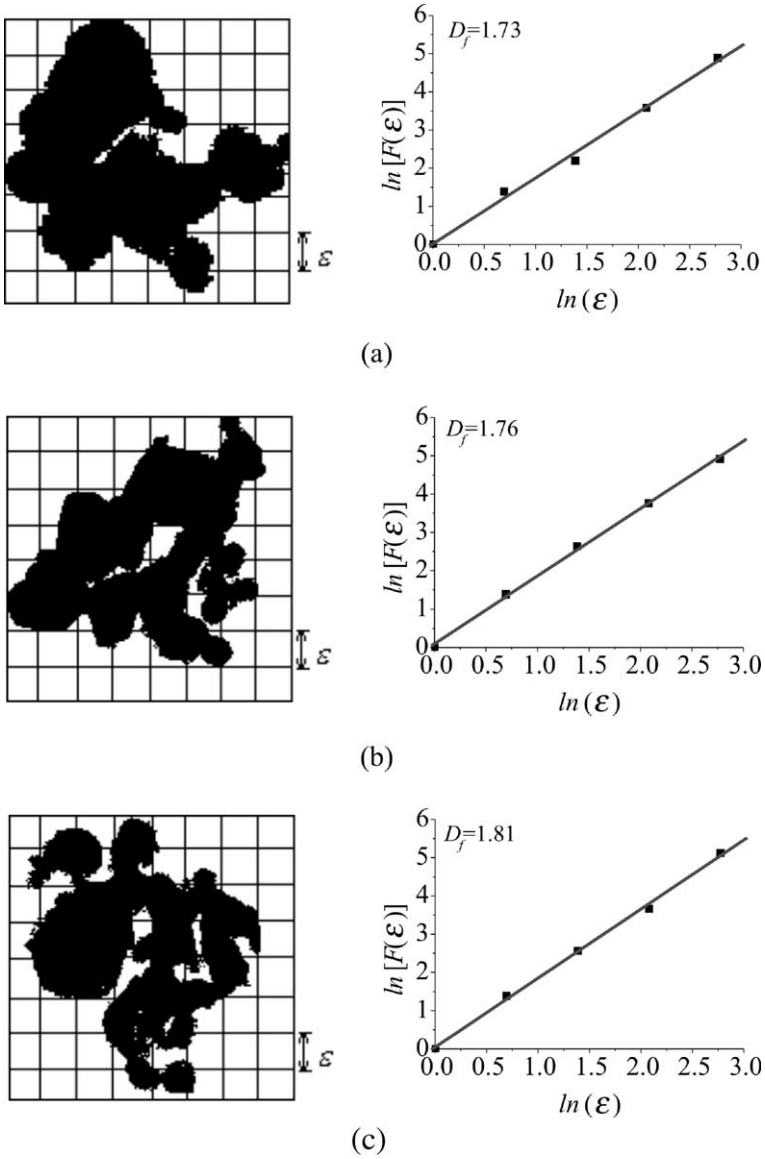

Fig. 5. The fractal dimension, D_f, of clustering CuO nanoparticles in H_2O. (a) Mass concentration = 0.02% ($\phi = 0.13\%$); (b) mass concentration = 0.04% ($\phi = 0.25\%$); (c) mass concentration = 0.06% ($\phi = 0.38\%$).

temperature difference between the heating surface and insulated bottom surface at quasi-steady-state, corresponding to Fourier number greater than 0.55. The cylindrical container for testing medium is 160 mm inside diameter and 9 mm deep. The Rayleigh number, Ra, of the medium being tested is controlled less than 10^3, so that liquid convection could be actually neglected. The estimated uncertainty for measured value of k_{eff} is within $\pm 2.9\%$. Our experimental set-up checked well with the measurements for thermal conductivity of liquids for deionized water and ethanol, i.e., $\phi = 0$, at temperature around 300 K. The testing specimen with CuO nanoparticles inclusion, $\phi > 0$, were prepared by applying supersonic wave for long time and no visible sediment was found in experiment. Besides, we added sodium dodecyl benzene sulphonate (SDBS), 2% by mass fraction, as the dispersion agent to further improve the distribution of particles in deionized water, and thus, to avoid the direct contact of CuO particles.

As a trial, we take the fractal dimension as $D_{\text{f1}} = 1.66$ and $D_{\text{f2}} = 1.57$ from Figs. 1 and 2. The calculated results with proposed fractal model were plotted and compared with the experimental data in Fig. 4. It is to be aware that these values of fractal dimension were derived from electron microscopic photos of clustering SiO_2 nanoparticles (25 nm in diameter) in ethanol. However, the predicted effective thermal conductivities still reflect qualitatively the tendency of variation if $\phi < 0.5\%$. The discrepancy may exist also due to the fact that anomalous diffusive index represents only the local fractal characteristic of particles suspensions.

To avoid or decrease such local effect, an alternative method is used, which assumes that the cluster distribution characteristics can be decided by the log normal function. Transmission electron microscopy (TEM) photos of 50 nm CuO particle suspensions in deionized water and the derived value of D_{f} for 50 nm CuO nanoparticle with mass concentration of 0.02%, 0.04% and 0.06% (corresponding to volume fraction of 0.13%, 0.25% and 0.38%), respectively, are quoted in Fig. 5. As shown, the fractal dimensions of clusters, D_{f}, for $\phi = 0.38\%$ is comparatively increased much more than that for $\phi = 0.25\%$.

Fig. 6 shows the calculated results by the fractal model proposed using the log normal function for their radius distribution. The modified fractal model fits well with experimental data when the particle concentration is less than 0.5%. Beyond this dilute limit, the possible deposition effect may be considered, which is difficult for the prediction of transport coefficients such as thermal conductivity.

As to compare the effect of adsorption on the nanoparticle surface, results with and without consideration of adsorption effect are both shown in Fig. 6. An obvious decrease in $k_{\text{eff}}/k_{\text{f}}$ is observed if the adsorption effect is not to be considered, and thus, the packed liquid molecules on the nanoparticle surfaces contribute obviously to the enhancement of effective thermal conductivity of liquid.

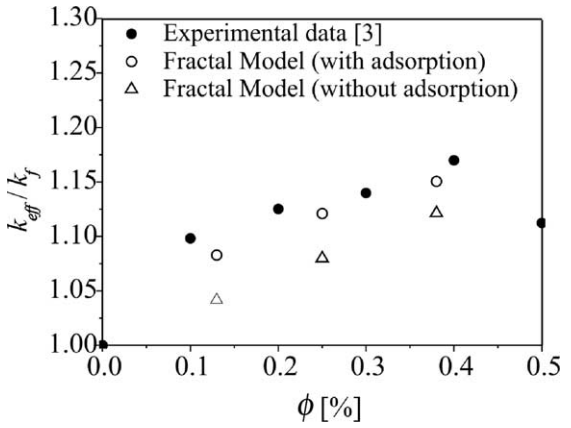

Fig. 6. Comparison of predicting results using fractal model and experimental data.

Though we predicted successfully the effective thermal conductivity of nanoparticle suspensions, the predictive calculation is complicated to involve the application and the improvement of the effective medium theory. Also, the space distribution of nanoparticle clusters should be carefully concerned and described. In addition, the prediction of effective thermal conductivity of adsorption monolayer needs to be further studied. In short, further research work would be needed to refine the model we proposed here, especially for suspension of metallic nanoparticles.

7. Conclusions

A fractal model is proposed for predicting the effective thermal conductivity of liquid with dilute suspension of nonmetallic nanoparticles. It involves the application and improvement of the effective medium theory.

The proposed fractal model predicts well the trend for variation of the effective thermal conductivity with dilute suspension of nanoparticles, and fits successfully with our experimental data for 50 nm CuO particles suspension in deionized water when $\phi < 0.5\%$. The calculated result also shows that the predictive calculation of effective thermal conductivity is complicated. Further work would be needed, especially for metallic nanoparticles inclusion.

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